# Dynamics of wrinkles on a vesicle in external flow

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Recent experiments by Kantsler et. al. (2007) have shown that the relaxational dynamics of a vesicle in external elongation flow is accompanied by the formation of wrinkles on a membrane. Motivated by these experiments we present a theory describing the dynamics of a wrinkled membrane. Formation of wrinkles is related to the dynamical instability induced by negative surface tension of the membrane. For quasi-spherical vesicles we perform analytical study of the wrinkle structure dynamics. We derive the expression for the instability threshold and identify three stages of the dynamics. The scaling laws for the temporal evolution of wrinkling wavelength and surface tension are established and confirmed numerically.

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Wrinkling of different thin sheets is a well-known effect which can be frequently observed in everyday life. Usually wrinkle patterns appear due to the external tensions applied to some material, or as a result of compression of inextensible films. Main properties of steady and/or equilibrium wrinkling structures are now well understood [1, 2]. However, much less is known about the dynamics of wrinkle structures. In certain biologically motivated experiments on membranes and vesicles [3, 4], the wrinkles are formed because of the instability induced by negative membrane tension, which is closely related to a buckling instability. Wrinkles formed this way exhibit non-trivial growth and relaxational dynamics. Theoretical description of these essentially non-equilibrium processes is a challenging problem which we attempt to approach in this paper. Although in this letter we focus on the analysis of the recent experiments [4], some predictions of our theory are universal and may be successfully applied to other systems where the formation of wrinkles is caused by negative tension.

Vesicles exhibit a variety of different regimes of motion in external fluid flows. These regimes were extensively studied during the last decade both by the experimentalists [5, 6, 7, 8] and theoreticians [9, 10, 11, 12, 13, 14, 15]. Depending on the external parameters a vesicle in external shear flow can experience three types of behavior: tank-treading, tumbling and trembling (also referred as vacillating-breathing [13] or swinging [12]) [8]. Although these motions correspond to quite non-trivial dynamics, the shape of a vesicle always remains smooth and can be effectively approximated by an ellipsoid. Recently, experiments performed by Kantsler et. al. [4] revealed a qualitatively new effect observed in non-stationary elongation flows. It was shown that in strong flows the relaxational dynamics of a vesicle is accompanied by the excitement of high order membrane deformation modes called wrinkles. Such dynamics can not be described in a framework of low-dimensional models which were used for the analysis of vesicle tank-treading, tumbling, and trembling. In this letter we extend these models to include the interaction between the vesicle shape and the wrinkle structure.

Before proceeding further we would also like to mention the experimental [16] and theoretical [17] investigations of wrinkle formation on microcapsules in external shear flows. Although this effect is similar to the one discussed here the underlying physics and main properties of wrinkles are essentially different. For example, the wrinkles which are observed on vesicles are not stationary and are excited only for a limited amount of time.

This letter is organized as follows: First we discuss the main features of a vesicle and show that the negative tension leads to instabilities of the flat membrane. Second, we present a model of a quasi-spherical vesicle in external flow. We derive the threshold of the instability and analyze the dynamics of wrinkle formation in strong flows. We show that one can distinguish three different stages of the dynamics. The first one lasts for a vanishingly small amount of time and is characterized by the exponential growth of short wavelength excitations. During the second stage the amplitude of wrinkles and their characteristic wavelengths grow algebraically. We derive the corresponding exponents and compare them to the experimental results. During the third stage the amplitude of wrinkles gradually reaches maximum and afterwards decays to zero. We find the scaling estimations for the wrinkle wavelength and the surface tension. In the end we present the results of our numerical simulations which confirm the analytical theory. We conclude by discussing the future challenges.

Vesicles are closed lipid bilayers which are incompressible and impermeable to the surrounding fluid. These two properties result in the conservation of the vesicle volume and the membrane area. Thus any vesicle is characterized by its excess area $\Delta$ which is the measure of vesicle's "nonsphericity": $A = (4\pi + \Delta)R^2$ where $A$ is the membrane area and $R$ is the effective vesicle radius, defined by the vesicle volume: $V = 4\pi R^3/3$. Free energy of the closed membrane is defined by the Helfrich functional and consists of the contributions from the bending energy and the surface tension $\sigma$ [18]:

$$F = \int dA \left( \frac{\kappa}{2} H^2 + \sigma \right). \qquad (1)$$

Here $H$ is the local mean curvature and $\kappa$ is the bending rigidity of a membrane. Note that for closed membrane geometry, the surface tension $\sigma$ is a quantity adjusting to other membrane parameters (similar to the pressure in an incompressible fluid) to ensure a given value of the membrane area $A$. It is useful to analyze the stability of a flat membrane with a given value of $\sigma$ before proceeding to the case of a closed membrane with a fixed area. Small perturbations of a flat membrane can be parameterized by a height function $z = h(x, y)$ which can be expanded in Fourier harmonics: $h(\mathbf{r}) = \sum h_{\mathbf{k}} \exp(i\mathbf{kr})$. The quadratic part of the Helfrich energy has the following form:

$$F = \frac{1}{2}\sum_{\mathbf{k}}(\kappa k^4 + \sigma k^2)|h_{\mathbf{k}}|^2. \qquad (2)$$

For positive $\sigma > 0$, this function is minimized by $u = 0$, so the flat membrane state is stable. However, for negative tension $\sigma < 0$, when the membrane is being shrinked, the modes with $k < \sqrt{|\sigma|/\kappa}$ become unstable. As we will show, this particular kind of instability is responsible for the formation of wrinkles in the experiment [4]. More precisely, we will show that the surface tension becomes negative when the direction of the external flow is reversed instantly. In this case small thermally induced membrane deformations on top of a smooth vesicle shape are amplified and lead to the formation of wrinkles.

In order to study this effect quantitatively we analyze the model of quasi-spherical ($\Delta \ll 1$) vesicles which proved itself to be very successful in analytical investigations of vesicle dynamics in external flows [9, 13, 14, 15]. The vesicle shape is parameterized by the small displacement function $u(\theta, \phi)$: $r = R(1 + u)$ which can be expanded in the series of spherical harmonics:

$$u(t, \theta, \phi) = \sum_{lm}\left[\frac{2\Delta}{(l-1)(l+2)}\right]^{1/2} u_{lm}(t)\mathcal{Y}_{lm}(\theta, \phi) \qquad (3)$$

The external velocity is assumed to have a linear elongational profile with the time-dependent strain: $\partial_x V_y = \partial_y V_x = 11\sqrt{5}/(16\sqrt{6\pi}) \cdot S(t)\sqrt{\Delta}/\tau$, where $\tau = \eta R^3/\kappa$ is the characteristic time-scale associated with the membrane bending forces. The numerical factor in this definition was included to simplify the expressions below. Throughout the paper we will discuss the experimentally interesting situation when $S(t)$ is the Heaviside-like function $S(t) = -S\mathrm{sign}(t)$.

The dynamical equations describing the dynamics of a quasi-spherical vesicle in external flow were first derived in [9] in the leading order in the small parameter $\sqrt{\Delta} \ll 1$:

$$\tau \dot{u}_{lm} = S(t) f_{lm} - (A_l\sigma + \Gamma_l)u_{lm} + \zeta_{lm}(t), \qquad (4)$$

Here $f_{lm} = \delta_{l,2}(\delta_{m,2} + \delta_{m,-2})$ and $\sigma(t)$ is the dimensionless angularly averaged part of the surface tension which is a Lagrangian multiplier associated with the excess area conservation constraint $\sum |u_{lm}|^2 = 1$. The numerical coefficients of (4) are given by: $\Gamma_l = (l-1)l^2(l+1)^2(l+2)/(2l+1)(2l^2+2l-1)$, and $A_l = l(l+1)(l^2+l-2)/(2l+1)(2l^2+2l-1)$. The statistical properties of thermal Langevin forces $\zeta_{lm}(t)$ can be found in [9]. In this letter we assume that the temperature $T$ is small, so that $\zeta_{lm}(t)$ in (4) are negligible during the evolution of the wrinkle structure. They only affect the dynamics through the non-zero initial conditions $u_{l,m}(t = 0)$ which ensure that the instability can indeed take the system out of the unstable state. In stationary planar elongational flows most of the excess area is stored in the $\mathcal{Y}_{2,\pm 2}$ harmonics. We therefore use the following parametrization : $u_{22} = u^*_{2,-2} = U$ and $u_{lm} = \sqrt{1 - |U|^2}\, w_{lm}$ for $l, m \neq 2, \pm 2$. The fraction of total excess area stored in the second-order spherical harmonics $\mathcal{Y}_{2\pm 2}$ is given by $|U|^2$. The argument of $U$ is related to the vesicle orientation angle $\Phi$ in the $xy$-plane: $U = |U|\exp(-2i\Phi)$. The normalization condition can be rewritten as $\sum |w_{lm}|^2 = 1$. The dynamical equations acquire the following form:

$$\tau\dot{U} = S(t) - (A_2\sigma + \Gamma_2)U \qquad (5)$$

Using the excess area conservation law one can also find the expression for the surface tension:

$$\sigma = \frac{S(t)\,\mathrm{Re}\,U - \Gamma_2|U|^2 - \bar{\Gamma}(1 - |U|^2)}{A_2|U|^2 + \bar{A}(1 - |U|^2)} \qquad (6)$$

where $\bar{A} = \sum A_l|w_{lm}|^2$ and $\bar{\Gamma} = \sum \Gamma_l|w_{lm}|^2$. Using the expression (6) one can easily show that there will be an instability for large enough values of $S$. Indeed, for constant positive $S(t) = S$ at $t < 0$ the vesicle exhibits small thermal fluctuations near the stable point $U = 1$. However as $S(t)$ changes sign this state becomes unstable and the vesicle starts rotating to the new stable point which corresponds to $U = -1$. The stability of the membrane can be studied by analyzing the expression (6). The surface tension instantly becomes negative after changing the velocity field: $\sigma(t = +0) = -(\Gamma_2 + S)/A_2$. Negative $\sigma$ can destabilize the high-order harmonics. The explicit condition can be found from (4). All harmonics of order up to $l$ become unstable if $A_l\sigma + \Gamma_l < 0$, which yields $S > S_l = A_2\Gamma_l/A_l - \Gamma_2$. For large $l \gg 1$ one can use the expansions $A_l \sim l/4, \Gamma_l \sim l^3/4$ to obtain $S_l \sim A_2 l^2 \gg 1$. The most unstable mode can be found by maximizing the growth increment $-\Gamma_l - A_l\sigma$. One obtains $l_0 = \sqrt{S/3A_2}$ for strong flows with $S \gg 1$.

Below the instability threshold, for relatively small values of $S$ higher order harmonics are not excited and the dynamics of a vesicle can be well described in terms of a single $U$ variable. The conservation of excess area implies that the dynamics is purely rotational and $|U|^2 = 1$. The characteristic time-scale associated with the rotational dynamics is estimated as $\tau/S$.

Describing the dynamics of the vesicle above the instability threshold is a considerably more difficult problem which requires an analysis of a complex nonlinear system (4) with large number of degrees of freedom. Fortunately, it is possible to approach this problem analytically

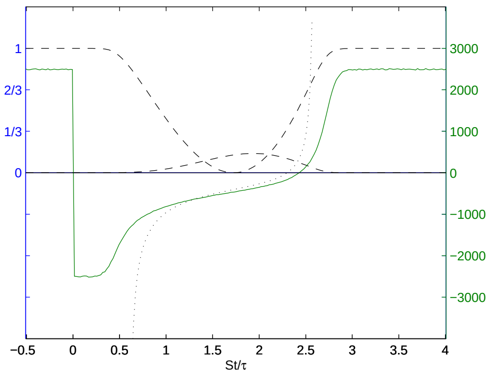


FIG. 1: Temporal dynamics of $|U|^2$, $\Delta_{19}$ (left grid) and surface tension $\sigma$ (right grid). Dotted line corresponds to the theoretical prediction (8).

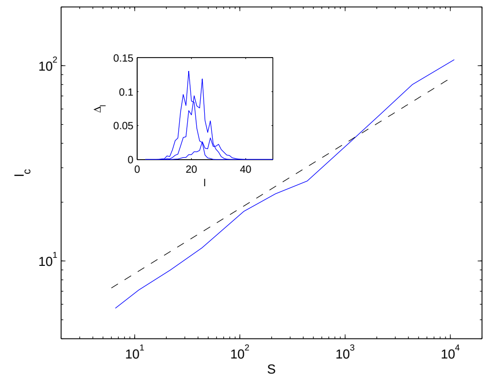


FIG. 2: $l_*$ versus the strain $S$. Linear curve corresponds to the scaling $l_* \propto S^{1/3}$. Inset: Distribution of $\Delta_l$ as a function of $l$ at the moments $tS/\tau = .7 : 1.3 : 2.2$ (right to left).

for strong flows which correspond to $l_0 \gg 1$. Although quantitative predictions of such an analysis can be applied only to strong flows, the qualitative picture is in most aspects the same even for moderate strains $S \gtrsim 1$.

For large values of $S$ one can distinguish between several different stages in the wrinkle evolution. At the first stage most of the excess area is stored in the second order angular harmonics and the surface tension can be assumed to be constant $\sigma = -2S/A_2$. Unstable higher order harmonics grow exponentially and the distribution of $w_{lm}$ becomes centered near the most unstable modes with $l = l_0 \sim \sqrt{S}$, so the terms $\bar{A}, \bar{\Gamma}$ can be estimated as $\bar{A} \sim \sqrt{S}$ and $\bar{\Gamma} \sim S^{3/2}$. This rapid growth saturates when the total excess area, stored in higher-order harmonics, becomes large enough so that their contribution to the surface tension becomes comparable to the contribution from the external flow. Formal condition can be found from (6): $(1 - |U|^2) \sim S^{-1/2} \ll 1$. Note that at this time most of the excess area is still stored in the second-order harmonics. The characteristic duration of this stage can be estimated as $\tau S^{-3/2}$. This time is much less than the characteristic relaxation time of the variable $U$, which can be estimated as $\tau S^{-1}$ from the equation (5) as in the situation below the instability threshold.

Second stage starts at $t \gtrsim \tau S^{-3/2}$ after the exponential growth has saturated. During the second stage higher order harmonics $w_{lm}$ provide the main contribution to the surface tension (6) which can be approximated as $\sigma = -(\bar{\Gamma}/\bar{A})$. Note that it depends only on the distribution of excess area over the higher-order harmonics $w_{lm}$. In order to find this distribution for large $l \gg 1$ one can use the formal solution of (4): $w_{lm}(t) = C w_{lm}(0) \exp\left(-\Gamma_l t/\tau + A_l \rho\right)$ where $C$ is the normalization constant and $\rho = -\int_0^t dt' \sigma(t')/\tau$. The distribution of excess area $\Delta_l = (1-|U|^2) \sum_m |w_{lm}|^2$ is a narrow function of $l$ centered around some $l_*$ which is determined as maximum of $A_l \rho - \Gamma_l t/\tau$: $l_* = \sqrt{\rho\tau/3t} \sim \sqrt{S}$. The characteristic width of the distribution can be also estimated in a usual way: $\delta l \sim (l_* t/\tau)^{-1/2}$. It is much less than $l_*$ for $t \gg \tau S^{-3/2}$. Narrowness of the distribution allows one to find the exact expression for the surface tension: $\sigma = -\bar{\Gamma}/\bar{A} = -l_c^2 = -\rho\tau/3t$. Using the definition of $\rho$ we obtain the following closed equation:

$$\dot{\rho} = \rho/3t \tag{7}$$

One can find its solution using the initial condition for the surface tension: $\sigma \sim \sqrt{S}$ at $t \sim \tau S^{-3/2}$. This yields $\rho = c(t/\tau)^{1/3}$, where $c$ is a constant of order unity. Similarly we obtain $\sigma = -(c/3)(t/\tau)^{-2/3}$ and $l_* = \sqrt{c/3}(t/\tau)^{-1/3}$. Therefore, second stage of the dynamics is characterized by the algebraic decay of the surface tension and by the narrow spectral distribution $\Delta_l$ of the wrinkling structure, whose peak smoothly drifts towards small $l$. As the absolute value of $l_*$ goes down the external velocity contribution $S(t)\,\mathrm{Re}\,U$ to the surface tension in (6) becomes important again. Comparing different contributions in (6) one can estimate the duration of the second stage as $\tau/S$. In the end of the second stage the peak of the $\Delta_l$ distribution is centered near $l_* \sim S^{1/3}$. This scaling law relates wavelength of wrinkles to the strain $\lambda = RS^{-1/3}$ and is one of the main results of this letter. Note that this wavelength is much smaller compared to the wavelength of the initially most unstable mode which is given by $RS^{-1/2}$. Our scaling is in reasonable agreement with the experimental results of Kantsler et. al. [4].

Second stage is followed by third one during which the surface tension in (6) is determined both by the external velocity and the wrinkles. The characteristic value of the surface tension can be estimated as $S^{2/3}$ which is much smaller than its initial value $\sigma(0) \sim S$. Due to a significant decrease of the surface tension, the dynamics of the second-order harmonics during the second stage is determined solely by the external velocity term $S(t)$ in (5). Therefore one can find the solution of (5) in the leading order: $U(t) = 1 - St/\tau$. The characteristic amplitude of wrinkles obeys the following simple law: $\sqrt{1 - |U|^2} = \sqrt{(2 - St/\tau)St/\tau}$. In order to analyze the

evolution of the surface tension $\sigma$ and the wrinkles wavelength $\lambda \sim R/l_*$ one has to solve the equation $\tau\dot{\rho} = \sigma$ keeping all the contributions to the surface tension. With the parametrization $\rho(t) = S^{-1/3} f(St/\tau)$ the problem is reduced to the ordinary differential equation:

$$\frac{df(x)}{dx} = \frac{f(x)}{3x} + 4\frac{1-x}{2-x}\sqrt{\frac{3}{xf(x)}} \tag{8}$$

with the initial condition $x^{-1/3} f(x) = c$ for $x = S^{-1/2}$ determined by the second stage. It is possible to solve (8) analytically: $f(x) = x^{1/3}\left[\log C\sqrt{S}x(2-x)\right]^{2/3}$, where $C$ is a constant of order unity. Note, that this solution adds only logarithmical factors of order $\log S$ to the previously derived scalings $l_* \sim S^{1/3}$ and $\sigma \sim S^{2/3}$. Although the second stage is formally present in the solution of (6) and corresponds to the limit $x \ll 1$ we purposely separate it from the third one because of its universality: during the second stage the surface tension is determined solely by the wrinkle structure. Therefore one may expect that the scaling laws $l_* \propto t^{-1/3}$ and $\sigma \propto t^{-2/3}$ will also hold for other problems where the initial negative surface tension was caused by different means.

We have tested our results by direct numerical simulations of equation (4). The total number of harmonics in our simulations was $l_{max} \approx 2\sqrt{S}$. The temperature which determines the power of the thermal noises $\zeta_{lm}$ in (4) was taken to be $T = 10^{-4}\kappa$. On FIG. 1 one can see the results of simulations for $S = 10^3$. Three temporal stages with qualitatively different dynamics are clearly distinguishable. The behavior of $|U|^2$ is very close to quadratic and the dynamics of $\sigma(t)$ may be well fitted by the solution of (8). The scaling law $l_* \propto S^{1/3}$ was confimed by simulations at different $S$ (FIG. 2 )

In conclusion we compile a list of main results presented in this paper. Motivated by recent experiments [4], we studied the relaxational dynamics of a vesicle in an elongational flow. We showed that high order membrane deformation modes are excited by the negative surface tension induced by external flow. For quasi-spherical vesicles we have found an analytical expression for the instability threshold and analyzed the evolution of the wrinkle structure. We identified three stages of the dynamics. The first stage corresponds to $t \lesssim \tau S^{-3/2}$ and is characterized by the exponential growth of unstable high order harmonics with the characteristic scales of order $\lambda \sim RS^{-1/2}$. This rapid growth quickly saturates and is followed by the second stage. For $\tau S^{-3/2} \ll t \lesssim \tau/S$ the surface tension decays algebraically as $\sigma(t) \propto t^{-1/3}$ and the characteristic wavelength of wrinkles grows as $\lambda \propto t^{1/3}$. Characteristic amplitude of the wrinkles grows as $\sqrt{t}$. During the third stage which ends at $t = 2\tau/S$ wrinkle amplitude behaves like $\sqrt{(2 - St/\tau)St/\tau}$ and the characteristic wavelength can be estimated as $\lambda \sim RS^{-1/3}$.

Finally we would like to note, that the algebraic growth of the characteristic wrinkle wavelength although with different exponent $\lambda(t) \propto t^{1/4}$ was observed in the studies of the thin rods dynamics experiencing the buckling instability [19]. We believe that the algebraic growth of wrinkle wavelength is a universal property of the interface dynamics experiencing the buckling instability. Although in this letter we considered a particular experiment, we believe that the scaling laws for the wrinkle wavelength $\lambda(t)$ and for the surface tension $\sigma(t)$ derived for the second stage are universal and can be observed in other experiments where the total area of the interface is conserved and the wrinkle growth is initiated by the external forces leading to negative surface tension.

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